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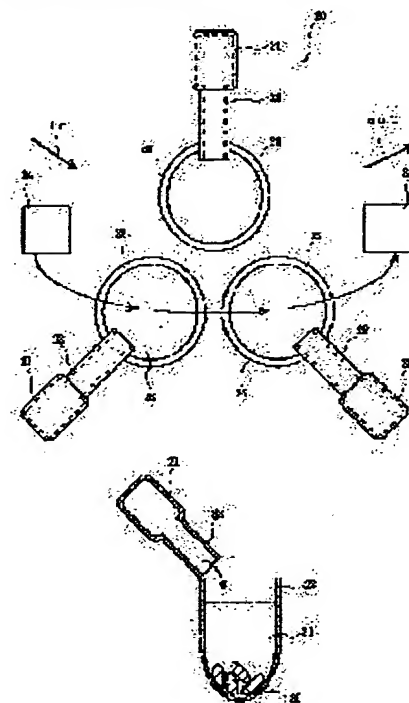
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(54) MANUFACTURE OF ELECTROLUMINESCENCE ELEMENT, ITS DEVICE, AND MANUFACTURE OF PELLET FOR ELECTROLUMINESCENCE ELEMENT

(57)Abstract:

PROBLEM TO BE SOLVED: To provide the manufacturing method of an electroluminescence element with high productivity capable of quickly supplying a constituting material of an organic layer of an organic EL element within a vacuum deposition device without contaminating the circumference, a device thereof, and the manufacturing method of a pellet for the electroluminescence element.

SOLUTION: Each material of an organic layer is formed in a pellet 25, the pellet 25 is housed in a holder 21, the holder 21 in which the pellet 25 of each material is housed is arranged on a crucible 23 placed within a vacuum chamber 20, the pellet 25 is supplied to the crucible 23 by spontaneous drop caused by gravity of the pellet 25, each pellet 25 is vaporized and deposited on an ITO substrate 24 carried on each crucible 23, and the organic layer is formed.



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CLAIMS

[Claim(s)]

[Claim 1] The manufacture method of electroluminescence devices that an organic layer including a luminescence field faces manufacturing the electroluminescence devices by which the laminating is carried out, pelletizes the component of the aforementioned organic layer, and forms the aforementioned organic layer physically using this pellet.

[Claim 2] The manufacture method of the electroluminescence devices indicated to the claim 1 using the pellet processed into the size of 1mm - several cm as the aforementioned pellet.

[Claim 3] The manufacture method of the electroluminescence devices indicated to the claim 1 which pays the raw material of the aforementioned component to a container with an open sand mold or a lid, carries out the heating dissolution, cools slowly below at sublimation temperature under a vacuum, and produces the aforementioned pellet.

[Claim 4] The manufacture method of the optical element indicated to the claim 1 which puts the aforementioned pellet into the container for vacuum evaporatio for every component kind of the aforementioned organic layer, and carries out vacuum deposition one by one.

[Claim 5] The manufacture method of the electroluminescence devices indicated to the claim 4 which holds the aforementioned pellet in an electrode holder, is made to carry out natural fall of the aforementioned pellet, and supplies it in the aforementioned container for vacuum evaporatio from this electrode holder.

[Claim 6] The manufacture method of electroluminescence devices optically indicated at the transparent base top to the claim 1 which carries out the laminating of a transparent electrode, an organic hole transporting bed, an organic luminous layer and/or an organic electronic transporting bed, and the metal electrode one by one.

[Claim 7] The manufacture method of the electroluminescence devices indicated to the claim 6 constituted as organic electroluminescence devices.

[Claim 8] The manufacturing installation of the electroluminescence devices characterized by having a supply means by which an organic layer including a luminescence field supplies the aforementioned pellet held in the equipment which manufactures the electroluminescence devices by which the laminating is carried out in a container, the electrode holder which holds the pellet of the component as the aforementioned organic layer, and this electrode holder in the aforementioned container.

[Claim 9] The manufacturing installation of the electroluminescence devices indicated to the claim 8 to which supply of the aforementioned pellet is performed by natural fall.

[Claim 10] The manufacturing installation of the electroluminescence devices indicated to the claim 8 by which is put into the aforementioned pellet for every component kind of the aforementioned organic layer by the container for vacuum evaporatio, and vacuum deposition is carried out one by one.

[Claim 11] The manufacturing installation of the electroluminescence devices indicated to the claim 8 using the pellet processed into the size of 1mm - several cm as the aforementioned pellet.

[Claim 12] The manufacturing installation of the electroluminescence devices indicated to the claim 8 by which the electroluminescence devices by which the laminating of a transparent electrode, an organic

hole transporting bed, an organic luminous layer and/or an organic electronic transporting bed, and the metal electrode was optically carried out one by one on the transparent base are manufactured optically. [Claim 13] The manufacturing installation of the electroluminescence devices indicated to the claim 12 by which organic electroluminescence devices are manufactured.

[Claim 14] The manufacture method of a pellet that face manufacturing the pellet used for the membrane formation of the aforementioned organic layer of electroluminescence devices by which the laminating is carried out, and an organic layer including a luminescence field carries out the heating dissolution, and pelletizes the raw material of the component of the aforementioned organic layer below at sublimation temperature under a vacuum.

[Claim 15] The manufacture method of a pellet which produces the aforementioned pellet in size of 1mm - several cm and which was indicated to the claim 14.

[Claim 16] The manufacture method of the pellet indicated to the claim 14 which pays the raw material of the aforementioned component to a container with an open sand mold or a lid, carries out the heating dissolution, cools slowly under a vacuum below at sublimation temperature, and produces the aforementioned pellet.

[Claim 17] It is the manufacture method of the pellet indicated to the claim 16 of using a container with the aforementioned lid when the aforementioned raw material has large sublimability.

[Claim 18] It is the manufacture method of the pellet indicated to the claim 16 of using the container of the aforementioned open sand mold when the aforementioned raw material has small sublimability.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the manufacture method of the manufacture method of electroluminescence devices, for example, the suitable electroluminescence devices for the organic electroluminescence display which is a flat-surface type display of spontaneous light, and uses an organic thin film for an electroluminescence layer, its equipment, and the pellet for electroluminescence devices.

[0002]

[Description of the Prior Art] In recent years, the importance of the interface of human being and a machine has been increasing by making multimedia-oriented goods into the start. In order for human being to do machine operation efficiently more comfortably, mistake the information from the machine operated, there is nothing, it is necessary to take out in sufficient amount briefly in an instant, and, for the reason, research is done about various display devices which make a display the start.

[0003] Moreover, the present condition is that the miniaturization of a display device and the demand to a thin shape are also increasing with the miniaturization of a machine every day.

[0004] For example, the miniaturization of the laptop type information management system which are display device one apparatus, such as a note type personal computer and a note type word processor, is astonishing, and the technical innovation about the liquid crystal display which is the display device also has a wonderful thing in connection with it.

[0005] The liquid crystal display is used as an interface of various products, and that for the product used our every day are used by making small television, a clock, and a calculator into the start not to mention the laptop type information management system today. [many]

[0006] These liquid crystal displays have been studied as a center of a display device as an interface of a machine with human being until they result in a small shell mass display device taking advantage of the feature that liquid crystal is a low-battery drive and a low power.

[0007] However, since this liquid crystal display needs big power rather than it needs a back light and drives liquid crystal to this back light drive, since there is no self-luminescence, as a result, by the built-in battery, a time becomes short and it has the limit on use.

[0008] Furthermore, since the angle of visibility of a liquid crystal display is narrow, it is also a big problem that it is not suitable for large-sized display devices, such as a large-sized display, and contrast changes with angles into an angle of visibility since it is the method of presentation by the orientation state of a liquid crystal molecule.

[0009] Moreover, although sufficient speed of response for the active matrix which is one of the drive methods to treat an animation if it thinks from a drive method is shown, in order to use a TFT drive circuit, enlargement of a screen size is difficult by the pixel defect. It is not desirable to use a TFT drive circuit even if it thinks from the point of a cost cut.

[0010] Although the passive matrix which is another drive method is a low cost and enlargement of a screen size is comparatively easy for it upwards, there is a problem that it does not have sufficient speed

of response to treat an animation.

[0011] On the other hand, as for the self-luminescence display device, a plasma display device, inorganic electroluminescence devices, organic electroluminescence devices, etc. are studied.

[0012] A plasma display device is what used plasma luminescence in low voltage gas for the display, and although it is suitable for enlargement and large capacity-ization, it has the problem in the field of thin-shape-izing and cost. Moreover, the AC bias of the high voltage is needed for a drive, and it is not suitable for a portable device.

[0013] Although the green luminescence display etc. was commercialized, with it being the same as that of a plasma display device, inorganic electroluminescence devices are AC-bias drives, and are considered for full-color-izing to be difficult by hundreds of drive required.

[0014] On the other hand, since the luminescence by carrier pouring to the anthracene single crystal which will generate fluorescence strongly at the first half of the 1960s was discovered, the electroluminescence phenomenon by the organic compound has been performed as fundamental research called carrier pouring to an organic material, since it was low brightness and monochrome and was moreover a single crystal, a long period and, although it had inquired.

[0015] However, it will be Eastman Kodak in 1987. Since Tang and others of a shrine announced the organic thin film electroluminescence devices of the laminated structure which has the amorphous luminous layer in which a low-battery drive and high brightness luminescence are possible, research and development of luminescence of R, G, and B in three primary colors, stability, brightness elevation, a laminated structure, the production method, etc. have been done briskly in every direction.

[0016] Furthermore, although it is the feature of an organic material, various new material is invented by the molecular design etc. and the application study to the color display of the organic electroluminescence display device which has the feature which was [luminescence / self-/ a direct-current low-battery drive, a thin shape,] excellent is also beginning to be performed briskly.

[0017] Organic electroluminescence devices (an organic EL element may be called hereafter) have the feature ideal as a spontaneous light [, such as transforming electrical energy into a light energy and emitting light in the shape of a field,] type display device by pouring in current.

[0018] Drawing 12 shows an example of conventional organic EL element 10. This organic EL element 10 produces the ITO (Indium tin oxide) transparent electrode 5, the hole transporting bed 4, a luminous layer 3, the electronic transporting bed 2, and cathode (for example, aluminum electrode) 1 one by one by the vacuum deposition method on the transparent substrate (for example, glass substrate) 6.

[0019] And by impressing direct current voltage 7 alternatively between the transparent electrodes 5 and cathode 1 which are an anode plate The electron into which the hole as a carrier poured in from the transparent electrode 5 was poured from cathode 1 through the hole transporting bed 4 reaches a luminous layer 3 through the electronic transporting bed 2, respectively, the reunion of an electronic-hole arises, the luminescence 8 of predetermined wavelength arises from here, and it can observe from the transparent substrate 6 side.
 [0020] In a luminous layer 3, you may use together the anthracene which are other photogenes, naphthalene, a phenanthrene, a pyrene, a chrysene, a perylene, a butadiene, a coumarin, an acridine, a stilbene, etc. The electronic transporting bed 2 can be made to contain the mixture with such fluorescence matter etc.

[0021] Drawing 13 shows another conventional example, in this example, it omits a luminous layer 3, makes the electronic transporting bed 2 contain mixture with the above-mentioned fluorescence matter, and shows organic EL-element 10A constituted so that the luminescence 8 of predetermined wavelength might arise from the interface of the electronic transporting bed 2 and the hole transporting bed 4.

[0022] Drawing 14 shows the example of the above-mentioned organic EL element. That is, although the layered product of each organic layer (the hole transporting bed 4, a luminous layer 3, or electronic transporting bed 2) is allotted between cathode 1 and an anode plate 5, these electrodes are made to cross in the shape of a matrix, and it prepares in the shape of a stripe, and a signal level is impressed to time series by the luminance-signal circuit 40 and the control circuit 41 with a built-in shift register, and it constitutes so that light may be made to emit in much intersection positions (pixel), respectively.

[0023] Therefore, of course, it becomes usable also as a picture reproducer as a display by such

composition. In addition, the above-mentioned stripe pattern can be arranged for every color of red (R), green (G), and blue (B), and it can constitute as full color or an object for multicolor.

[0024] Generally, the organic thin film layers 2, 3, and 4 which emit light in the display device which used such an organic EL element, and which consists of two or more pixels are pinched between the transparent electrode 5 and the metal electrode 1, and emit light to a transparent-electrode 5 side.

[0025] However, the field also with the above, organic, still more nearly unsolved EL elements exists.

[0026] That is, luminescence by which RGB three primary colors were stabilized when performing application to the color display of an organic EL element is indispensable conditions. However, in the organic EL element of RGB three primary colors, since the electronic states which the luminescent material of each color has differ, it is thought that the highest occupancy molecular orbital (HOMO) of luminescent material differs also from minimum non-occupying molecular orbital (LUMO) level.

[0027] This shows that the optimal structure in the organic stacked-volume layer of each color changes with colors, and it is shown that the optimal material for luminous layers which sandwich a luminous layer, such as a hole transporting bed and an electronic transporting bed, differs in each color.

[0028] Moreover, the laminating of two or more layers is carried out in fact, and what is considered as plans, such as an improvement of the luminous efficiency of an organic EL element and reinforcement, is used, although light can be theoretically emitted also by the monolayer. Especially a hole pouring layer (buffer layer for pouring in a charge from an anode to an organic material) and a hole transporting bed (layer for pouring in a charge from a hole pouring layer efficiently to a luminous layer) are indispensable to a highly efficient organic EL element.

[0029] Therefore, if it is going to produce the organic EL element which has the optimized luminous layer of RGB three primary colors, the lamination of a different hole pouring layer, a hole transporting bed, a RGB luminous layer, and an electronic transporting bed is required, and 12 kinds of material is needed only in an organic layer.

[0030]

[Problem(s) to be Solved by the Invention] However, when it is necessary to produce by the multi-chamber process and application to a color display is performed for the improvement in a property of an organic EL element, considering realizing a low-battery drive by layer structure which is different in each color, an extensive improvement of a facility of the vacuum evaporation system which is production equipment will be needed, and it will become large-sized equipment actually.

[0031] Furthermore, although considering mass-production nature it is required to supply material quickly and simple, the organic material used for organic EL-element production is usually powder in many cases. Therefore, after carrying out air opening of the vacuum evaporation system, the case where exchange the vacuum evaporatio no boat containing the organic material, or pay [that an organic material does not disperse outside and] careful attention, and it adds directly into a crucible is almost the case.

[0032] However, in the conventional process like the above, it is easy to mix the dust and impurity from the outside, and dispersion and the yield of a property of a product in mass production are affected.

[0033] Therefore, although it is desirable to perform supply of material in the process of vacuum consistency quickly [it is quick and] if it can do without an organic material polluting the circumference in order to solve these problems, the method of effective material supply does not exist.

[0034] Then, the purpose of this invention has supply of material in offering the quick manufacture method of the manufacture method of the high electroluminescence devices of the productivity which can be performed in a vacuum process, without polluting the circumference, its equipment, and the pellet for electroluminescence devices.

[0035]

[Means for Solving the Problem] That is, an organic layer including a luminescence field faces manufacturing the electroluminescence devices by which the laminating is carried out, and pelletizes the component of the aforementioned organic layer, and this invention requires it for the manufacture method (the manufacture method of this invention is called hereafter.) of electroluminescence devices which forms the aforementioned organic layer physically using this pellet.

[0036] The pelletized material which can also supply surrounding contamination quickly nothing and does not have an impurity by the operation like sublimation refining in the process of pellet production or removal of a remains solvent can be used without dispersing at the time of the supply, since an organic layer is formed using the pelletized component according to the manufacture method of this invention. Therefore, while the electroluminescence devices which have the organic layer by which the property was stabilized are obtained, material supply can be performed in a vacuum using a pellet supply means, and electroluminescence devices can be manufactured with productivity sufficient at the multi-chamber process.

[0037] Moreover, this invention requires an organic layer including a luminescence field for the manufacturing installation (the manufacturing installation of this invention is called hereafter.) of the electroluminescence devices characterized by having a container, the electrode holder which holds the pellet of the component as the aforementioned organic layer, and a supply means to supply the aforementioned pellet held in this electrode holder in the aforementioned container in the equipment which manufactures the electroluminescence devices by which the laminating is carried out.

[0038] According to the manufacturing installation of this invention, a pellet can be held in an electrode holder, a container can be supplied, and a manufacturing installation with the sufficient repeatability of the above-mentioned manufacture method can be offered.

[0039] Moreover, an organic layer including a luminescence field faces this invention manufacturing the pellet used for the membrane formation of the aforementioned organic layer of electroluminescence devices by which the laminating is carried out, and it requires it for the manufacture method (the manufacture method of the pellet of this invention is called hereafter.) of a pellet which carries out the heating dissolution and pelletizes the raw material of the component of the aforementioned organic layer below at sublimation temperature under a vacuum.

[0040] According to the manufacture method of the pellet of this invention, since the heating dissolution of the raw material is carried out below at sublimation temperature under a vacuum, sublimation refining of the raw material is carried out, a remains solvent is removed in this process, and the manufacture method of the pellet of the organic material for electroluminescence devices which has the organic layer which consists of a pure material without a pollutant can be offered.

[0041] here -- setting -- "a vacuum" -- 10-3Torr (0.13Pa) -- further -- the state of a high vacuum is pointed out from 10-6Torr (1.3x10 to 4 Pa), or it

[0042]

[Embodiments of the Invention] Hereafter, the form of desirable operation of this invention is explained, referring to a drawing.

[0043] In the manufacture method of the manufacture method of the above-mentioned this invention, a manufacturing installation, and a pellet, although it is desirable to use a pellet with a size of 1mm - several cm which processed the raw material of an organic material as the aforementioned pellet, size is not limited to this.

[0044] It is desirable to put into a container with an open sand mold or a lid as shown in drawing 3 , to carry out the heating dissolution, to anneal the raw material of the aforementioned component below at sublimation temperature under a vacuum, in production of this pellet, and to produce the aforementioned pellet. Thereby, since a large material of sublimability adheres to the internal surface of a top cover 33 and a small material of sublimability melts and stops in the dissolution pan 31 where sublimation refining of the raw material was carried out and a remains impurity is removed, as shown in drawing 4 , this container 30 can be disassembled and pellets can be collected easily.

[0045] Therefore, aluminum quinoline complex Alq3 which a raw material shows to drawing 8 When it is desirable to use a container with a lid when sublimability is large like alpha-NPD shown in drawing 7 and sublimability is small like m-MTDATA which a raw material shows to drawing 6 , the container of an open sand mold can be used.

[0046] Moreover, things other than this are sufficient as a container, and it can devise internal structure and can form it arbitrarily so that the pellet of a fixed configuration may be obtained.

[0047] And as typically shown in drawing 1 , it is desirable to put the produced pellet into the container

23 for vacuum evaporation for every component kind of the aforementioned organic layer, to convey the substrate 24 in which ITO was formed within a chamber 20, and to carry out vacuum deposition one by one.

[0048] In this case, as shown in drawing 2, in order to hold a pellet in an electrode holder 21, to carry out natural fall of the aforementioned pellet into the aforementioned container 23 for vacuum evaporation from this electrode holder 21 and to form the specified quantity, it is desirable to supply the material of a complement.

[0049] By forming an organic layer using the pellet obtained as mentioned above, optically, on a transparent base, the laminating of a transparent electrode, an organic hole transporting bed, an organic luminous layer and/or an organic electronic transporting bed, and the metal electrode can be carried out one by one optically, and it can constitute as good organic electroluminescence devices.

[0050] Moreover, the organic EL element to produce may carry out the laminating of the ITO anode electrode 12, the hole pouring layer 13, the hole transporting bed 14, a luminous layer 15, the electronic transporting bed 16, and the cathode electrode 17 on a substrate 11, as shown in drawing 5, and especially this component is not limited. For example, if it is the hole transporting bed 14, hole transportation material, such as a benzidine derivative, a styryl amine derivative, a triphenylmethane-color derivative, and a hydrazone derivative, can also be used. Moreover, if the electronic transporting bed 16 is made to contain a photogene like drawing 13 mentioned already, and it can also be made the single hetero type with which the electronic transporting bed 16 made the luminous layer serve a double purpose and the metal electrode of a cathode is formed thinly, luminescence can also be taken out from a cathode side.

[0051] Moreover, the ITO anode electrode 12, the hole pouring layer 13, the hole transporting bed 14, a luminous layer 15, the electronic transporting bed 16, and the cathode electrode 17 may be laminated structures which each becomes from two or more layers.

[0052] Furthermore, vapor codeposition of a minute amount molecule may be performed for control of the emission spectrum of a luminous layer 15, for example, you may be a minute amount **** organic thin film about organic substances, such as a perylene derivative, a coumarin derivative, and pyran system coloring matter.

[0053] About the material of the cathode electrode 17, in order to pour in an electron efficiently, it is desirable to use the small metal of the work function from the vacuum level of an electrode material, it is a simple substance about low work function metals, such as In, Mg, Ag, calcium, Ba, and Li, or you may use it as an alloy with other metals, raising stability.

[0054] In the form of this operation, although ITO which is a transparent electrode is used for the anode electrode 12 in order to take out luminescence from an anode electrode side, in order to pour in a hole efficiently, of course, you may use electrodes, such as what has a large work function from the vacuum level of an anode electrode material, for example, Au, SnO₂+Sb, and ZnO+aluminum.

[0055] Furthermore, in order to raise stability, as an imaginary line shows, a protective coat may be prepared in drawing 5 with a germanium oxide etc., the influence of the oxygen in a closure deed and the atmosphere etc. may be eliminated, and an element may be driven in the state where it lengthened to the vacuum, of course.

[0056] Moreover, heat-treatment, sublimation processing, and an organic solvent may be used for the method of pelletizing, pellet size needs to be influenced by a property, a pelletizing art, etc. of material, in a small thing, although it becomes the shape of a several cm pellet from 1mm by the big thing, pellet size does not need to be single, for example, the several cm pellet may be intermingled from 1mm.

[0057]

[Example] Hereafter, the example of this invention is explained.

[0058] According to the form of desirable operation of this invention mentioned above, membrane formation equipment (a chamber may be called hereafter) was produced. Drawing 1 is the ** type view showing the composition superficially, and drawing 2 is some of these outline cross sections.

[0059] A pellet 25 is put into a crucible 26 for every material which constitutes an organic layer, it melts, and the number of material kinds required as a fusion material 26 is allotted, as shown in drawing

2, the electrode holder 21 which put in the pellet 25 for supply on the crucible 26 further is arranged, and the supply races 22 are formed successively at the nose of cam of an electrode holder 21, and it is formed in the configuration which is easy to supply a pellet to a crucible 26.

[0060] Drawing 1 shows that membranes are formed one by one and an organic layer is taken out to Arrow out side, when the state where 3 sets has been placed in a fixed position in one vacuum chamber (illustration abbreviation) in the group of such an electrode holder 21 and a crucible 23 is shown, and it is carried in a chamber, and the substrate 24 after forming an ITO transparent electrode from Arrow in side has the crucible 23 top for every material kind conveyed and goes round it.

[0061] A crucible 23 is supplied from an electrode holder 21 at each using the natural fall by gravity. The timing of supply establishes a calibration curve from the relation between the heating temperature (the amount of current supplied) of the crucible 23 according to each organic material, and an evaporation rate, the decrement of the material in a crucible 23 is estimated, and the feedback system is taken so that the melting-material 26 may always exist in a crucible 23.

[0062] Drawing 3 shows the container for producing a pellet, (a) is an outline cross section, (b) is a plan, and drawing 4 shows the cross section in the state where this container was disassembled.

[0063] Like illustration, this container 30 consists of dissolution **** 31, an inside lid 32, and a top cover 33, and Collars 31a, 32a, and 33a are installed, and if these collars 31a, 32a, and 33a pile up, it has structure which can carry out a seal from each rim. As a material of a container 30, a tantalum, molybdenum, or a tungsten can use it suitably.

[0064] The imaginary line shown in drawing 3 (a) and drawing 4 shows the pellet 25 under production. As shown in drawing 3 (a), the raw material 27 with large sublimability is paid into dissolution **** 31, closes the inside lid 32 and a top cover 33, and heats them by resistance heating.

[0065] Like illustration, the hole 34 is established in the inside [this] lid 32 at two places, and when sublimability is a large raw material, it is used in the state of drawing 3 (a). Therefore, once it heats and sublimates the heated large raw material of sublimability and it hits a top cover 33 from this hole 34, it comes out to the hole 35 shell exterior of a top cover 33. However, since it heats near the sublimation temperature, there are few amounts which come out to the exterior, and many adhere to internal-surface 33b of a top cover 33.

[0066] However, it is **** to the ***** dissolution pan 31, without sublimating, since it heats below at sublimation temperature, when sublimability is a small raw material. Therefore, only using the dissolution pan 31, even if the inside lid 32 and a top cover 33 do not use it, they can make a pellet.

[0067] Each production of the above-mentioned pellet is performed within the vacuum system (illustration ellipsis) according to a vacuum chamber or it. And drawing of the produced pellet can be easily taken out, if a container 30 is disassembled as shown in drawing 4. Although there is no limit in the degree of vacuum of pellet production, the degree of high vacuum can obtain the pellet of a high grade preferably.

[0068] Drawing 5 shows an example of the organic EL element which formed the pellet 25 mentioned above as a material of an organic layer, and carries out the laminating of the metal electrode 17 as the ITO transparent electrode 12, the hole pouring layer 13, the hole transporting bed 14, the luminous layer 15, the electronic transporting bed 16, and cathode as an anode one by one on a glass substrate 11.

[0069] And while forming the organic EL element using the pellet produced from the raw material like the example shown below as a component of the organic layer of this organic EL element, the organic EL element using the powdery raw material which is not pelletized was formed as an example of comparison, and both were compared.

[0070] <Example 1> m-MTDATA of the structure expression shown in drawing 6 as a hole-injection layer material in this example (4, 4', 4''-tris(3-methylphenylphenylamino) triphenylamine) It used. As shown in pelletizing processing at drawing 3, 1.0g of m-MTDATA was put into the dissolution pan 11 made from an open-sand-mold tantalum (the same is said of each following example) which uses neither a top cover nor an inside lid, and it became hot slowly until material dissolved in the vacuum. Dissolved m-MTDATA became a vitrified pellet, before the sublimation after dissolution occurred, it stopped heating, and it annealed it to the room temperature.

[0071] The existence of sublimation checked by the thickness sensor monitor. Pelletizing of this m-MTDATA was simultaneously performed using four containers 30 shown in drawing 3 . Thus, the produced m-MTDATA pellet turned into a pellet with a 20mmx10mmx thickness of about 5mm.

[0072] <Example 2> In this example, alpha-NPD (4 and 4'-bis[N-(1-naphthyl)-N-phenyl-amino] biphenyl) of the structure expression shown in drawing 7 was used as an electron hole transporting-bed material. Compared with m-MTDATA, sublimability of alpha-NPD is large, and it is difficult to pelletize by sublimation of material only in an open-sand-mold dissolution pan 11 like an example 1. Therefore, as pelletizing processing, as shown in drawing 3 , it carried out with the lid. 1.0g of alpha-NPD was put in into the state of drawing 3 , and in the vacuum, material became hot slowly until sublimation had been sensed by the thickness sensor monitor. And in the 1/100nm or less state, the evaporation rate stopped heating and cooled slowly to the room temperature.

[0073] Pelletizing of this alpha-NPD was simultaneously performed using four containers 30 like the example 1. Thus, the produced alpha-NPD pellet turned into a pellet with a 15mmx10mmx thickness of about 3mm, adhered to internal-surface 33b of a top cover 33, and was pelletized.

[0074] <Example 3> Alq3 of the structure expression shown in drawing 8 as an electronic transportability luminous layer material in this example (8-hydroxy quinorine aluminum) It used. Alq3 Compared with m-MDATA, sublimability is large, and pelletizing is difficult by sublimation of material like an example 2 in the dissolution pan 11 of an open sand mold. Therefore, it uses with a lid like an example 2 as pelletizing processing, and is Alq3 similarly. 1.0g was put in, and in the vacuum, material became hot slowly until sublimation had been sensed by the thickness sensor monitor. And in the state 1/100nm or less, the evaporation rate stopped heating and cooled slowly to the room temperature.

[0075] This Alq3 Pelletizing was simultaneously performed using four containers 30 like the example 1. Thus, produced Alq3 The pellet turned into a pellet with a 15mmx10mmx thickness of about 3mm, adhered to internal-surface 33b of a top cover 33, and was pelletized.

[0076] <Example 4> In this example, an organic EL element like drawing 5 was produced using the pellet produced by the above-mentioned examples 1-3. As shown in drawing 10 , size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx glass substrate 11 which is 30mm SiO₂ The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. It is Alq3 considering alpha-NPD as 20nm and an electronic transporting bed 16 considering m-MTDATA as 30nm and a hole transporting bed 14 as a hole pouring layer 13 on this substrate. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation of aluminum was performed as a metal electrode 17 of a cathode.

[0077] Using the vacuum evaporation system 20 shown in drawing 1 , in early stages of vacuum evaporation, beforehand, production of this organic EL element put one pellet 25 of each material at a time into the crucible 23, and set it to it. As mentioned already after that, the substrate of a 30mm angle like drawing 10 was able to be produced at about 200 time [1] by supplying a pellet 25, without carrying out air opening of the chamber 20.

[0078] <Example 5> BSB-BCN of the structure expression shown in drawing 9 as a luminescent material in this example (2, 5-Bis[4-(N and N-4-methoxydiphenylamino) styryl]-1, 4-dicyanobenzene) It used. 1.0g of BSB-BCN was put into the dissolution pan 11 of an open sand mold like the example 1 as pelletizing processing, and it became hot slowly until material dissolved in the vacuum. Dissolved BSB-BCN became a vitrified pellet, before the sublimation after dissolution occurred, it stopped heating, and it annealed it to the room temperature.

[0079] The existence of sublimation checked by the thickness sensor monitor. Pelletizing of this BSB-BCN was simultaneously performed like the above-mentioned examples 1-3 using four containers 30 shown in drawing 3 . Thus, the produced m-MTDATA pellet turned into a pellet with a 20mmx10mmx thickness of about 5mm.

[0080] Next, the organic EL element which emits light in red like drawing 5 , using the pellet which consists of this BSB-BCN as a luminous layer was produced. As shown in drawing 10 , size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx

glass substrate 11 which is 30mm SiO₂ The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. m-MTDATA as a hole pouring layer 13 on this substrate 30nm, BSB-BCN as 30nm and a luminous layer 15 for alpha-NPD as a hole transporting bed 14 30nm, It is Alq₃ as an electronic transporting bed 16. The laminating of the 30nm was carried out one by one, and, finally the vacuum evaporation of AlLi (aluminum lithium alloy and Li concentration % of one mol) was performed as a metal electrode 17 of a cathode.

[0081] Using the vacuum evaporation system 20 shown in drawing 1 like the example 4, in early stages of vacuum evaporation, production of this organic EL element put one pellet 25 of each material at a time into the crucible 3 beforehand, and set it to it. As mentioned already after that, about 200 substrates of a 30mm angle like drawing 10 were able to be produced at once by supplying a pellet 25, without carrying out air opening of the chamber 20.

[0082] <Example 1 of comparison> In this example, it did not pelletize but an organic EL element like drawing 5 (the same is said of each following example of comparison) was produced using each organic powdered layer material. As shown in drawing 10, size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx glass substrate 11 which is 30mm SiO₂ The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. Powdered m-MTDATA, alpha-NPD, and Alq₃ It is made to correspond to the examples 1-3 which mentioned each material already. It puts in about g. the container of drawing 3 -- 0.5-0.7 -- m-MTDATA 30nm as a hole pouring layer 13 on this substrate It is Alq₃ considering alpha-NPD as 20nm and an electronic transporting bed 16 as a hole transporting bed 14. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation of aluminum as a metal electrode 17 of a cathode was performed.

[0083] However, by this method, by the time material was lost, the limitation carried out the vacuum evaporation of the about 20-30 substrates of a 30mm angle like drawing 10.

[0084] <Example 2 of comparison> It is Alq₃ considering alpha-NPD as 20nm and an electronic transporting bed 16 considering m-MTDATA as 30nm and a hole transporting bed 14 as a hole pouring layer 13 on the substrate produced like the above-mentioned example 1 of comparison in this example. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation of aluminum was performed as a metal electrode 17 of a cathode. By this method, although about 40 substrates production was [*****/ before supplying material] possible, there was 20-30 number of sheets which can produce after supply of material.

[0085] Therefore, in order to pursue this cause, when the material distribution system was checked, the powdered organic material adhered to the rail 22 for material supply shown in drawing 1 and drawing 2, and it remained, without falling into a crucible 23 completely. Moreover, the organic material might fall around. Furthermore, as compared with the device of an example 4, luminous efficiency was low, mixing of material etc. was suggested and the organic EL element produced after material supply had a bad yield.

[0086] <Example 3 of comparison> BSB-BCN of the structure expression shown in drawing 9 as a luminescent material in this example (2, 5-Bis[4-(N and N-4-methoxydiphenylamino) styryl]-1, 4-dicyanobenzene) Although used, it did not pelletize, but raw material was supplied in the state of powder, and the organic EL element of red luminescence was produced.

[0087] That is, it considers as the hole pouring layer 13 on the substrate produced like the above-mentioned example of comparison, and is Alq₃ considering BSB-BCN as 30nm and an electronic transporting bed 16 considering alpha-NPD as 30nm and a luminous layer 15 considering m-MTDATA as 30nm and a hole transporting bed 14. The laminating of the 30nm was carried out one by one, and, finally the vacuum evaporation of AlLi (aluminum lithium alloy and Li concentration % of one mol) was performed as a metal electrode 17 of a cathode.

[0088] Drawing 11 is the organic EL element shown in drawing 5, and is the graph of the data which compare the brightness and luminous efficiency the case where it uses with the material which pelletized

BSB-BCN of an example 5 as a luminous layer 15, and at the time of using the powder material of BSB-BCN shown in the example 3 of comparison as a luminous layer 15.

[0089] As shown in this graph, when it deposits with the material which pelletized BSB-BCN Brightness is about 8000 cd/m² at the time of applied-voltage 10.5V. When it is shown and deposits by powdered BSB-BCN to the luminous efficiency at this time being between 0.45 - 0.5 lm/W, brightness is cd [8000]/m² at 11V. Although it is almost the same Since luminous efficiency has become between 0.25 - 0.3 lm/W, in the case of a powdered material, it turns out that the property of an element is clearly inferior. That is, by pelletizing material, moisture, an impurity, etc. of a raw material are removed and it can be proved that it is an effect by the purity of material having been improved.

[0090] the material kind which constitutes the organic layer of an organic EL element according to this example -- since vacuum evaporatio no can be performed supplying the material moreover pelletized to the crucible 23 in the vacuum using the material pelletized to things, air opening of the vacuum evaporatio no chamber cannot be carried out because of vacuum evaporatio no material supply, and a lot of elements can be produced at once

[0091] Moreover, since the container for pellet production can be used irrespective of the size of the sublimability of material, it is efficient, and since it can decompose, drawing of the produced pellet is also easy.

[0092] Furthermore, by being also the head end process of vacuum evaporatio no material, consequently performing sublimation refining and removal of a remains solvent simultaneously, and using this pellet, the process of pelletizing is stabilized and, in other words, can produce the efficient device by which the property was stabilized with the sufficient yield.

[0093] The above-mentioned example can be deformed based on the technical thought of this invention.

[0094] For example, the vacuum evaporatio no equipment shown in drawing 1 can also be made into the composition and the configurations other than an example, and can be carried out arbitrarily [the supply method of the pellet of a crucible 23 shown in drawing 2] besides an example.

[0095] Moreover, the container 30 for pellet production shown in drawing 3 can also be made into the composition and the configurations other than an example. In order to standardize the configuration of the pellet produced especially in a fixed configuration, a diaphragm can also be formed in the internal surface of dissolution **** 31 and a top cover 33.

[0096]

[Function and Effect of the Invention] Since according to this invention carry out the heating dissolution, the component of an organic layer is pelletized below at sublimation temperature under a vacuum, this pellet is supplied to the container for vacuum evaporatio no using a supply means and the aforementioned organic layer is physically formed using this pellet as mentioned above The pelletized material which can also supply surrounding contamination quickly nothing and does not have an impurity by the operation like sublimation refining in the process of pellet production or removal of a remains solvent can be used without dispersing at the time of supply of the material. Therefore, while the electroluminescence devices which have the organic layer by which the property was stabilized are obtained, material supply can be performed in a vacuum using a pellet supply means, and electroluminescence devices can be manufactured with productivity sufficient at the multi-chamber process.

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TECHNICAL FIELD

[The technical field to which invention belongs] this invention relates to the manufacture method of the manufacture method of electroluminescence devices, for example, the suitable electroluminescence devices for the organic electroluminescence display which is a flat-surface type display of spontaneous light, and uses an organic thin film for an electroluminescence layer, its equipment, and the pellet for electroluminescence devices.

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PRIOR ART

[Description of the Prior Art] In recent years, the importance of the interface of human being and a machine has been increasing by making multimedia-oriented goods into the start. In order for human being to do machine operation efficiently more comfortably, mistake the information from the machine operated, there is nothing, it is necessary to take out in sufficient amount briefly in an instant, and, for the reason, research is done about various display devices which make a display the start.

[0003] Moreover, the present condition is that the miniaturization of a display device and the demand to a thin shape are also increasing with the miniaturization of a machine every day.

[0004] For example, the miniaturization of the laptop type information management system which are display device one apparatus, such as a note type personal computer and a note type word processor, is astonishing, and the technical innovation about the liquid crystal display which is the display device also has a wonderful thing in connection with it.

[0005] The liquid crystal display is used as an interface of various products, and that for the product used our every day are used by making small television, a clock, and a calculator into the start not to mention the laptop type information management system today. [many]

[0006] These liquid crystal displays have been studied as a center of a display device as an interface of a machine with human being until they result in a small shell mass display device taking advantage of the feature that liquid crystal is a low-battery drive and a low power.

[0007] However, since this liquid crystal display needs big power rather than it needs a back light and drives liquid crystal to this back light drive, since there is no self-luminescence, as a result, by the built-in battery, a time becomes short and it has the limit on use.

[0008] Furthermore, since the angle of visibility of a liquid crystal display is narrow, it is also a big problem that it is not suitable for large-sized display devices, such as a large-sized display, and contrast changes with angles into an angle of visibility since it is the method of presentation by the orientation state of a liquid crystal molecule.

[0009] Moreover, although sufficient speed of response for the active matrix which is one of the drive methods to treat an animation if it thinks from a drive method is shown, in order to use a TFT drive circuit, enlargement of a screen size is difficult by the pixel defect. It is not desirable to use a TFT drive circuit even if it thinks from the point of a cost cut.

[0010] Although the passive matrix which is another drive method is a low cost and enlargement of a screen size is comparatively easy for it upwards, there is a problem that it does not have sufficient speed of response to treat an animation.

[0011] On the other hand, as for the self-luminescence display device, a plasma display device, inorganic electroluminescence devices, organic electroluminescence devices, etc. are studied.

[0012] A plasma display device is what used plasma luminescence in low voltage gas for the display, and although it is suitable for enlargement and large capacity-ization, it has the problem in the field of thin-shape-izing and cost. Moreover, the AC bias of the high voltage is needed for a drive, and it is not suitable for a portable device.

[0013] Although the green luminescence display etc. was commercialized, with it being the same as that

of a plasma display device, inorganic electroluminescence devices are AC-bias drives, and are considered for full-color-izing to be difficult by hundreds of drive required.

[0014] On the other hand, since the luminescence by carrier pouring to the anthracene single crystal which will generate fluorescence strongly at the first half of the 1960s was discovered, the electroluminescence phenomenon by the organic compound has been performed as fundamental research called carrier pouring to an organic material, since it was low brightness and monochrome and was moreover a single crystal, a long period and, although it had inquired.

[0015] However, it will be Eastman Kodak in 1987. Since Tang and others of a shrine announced the organic thin film electroluminescence devices of the laminated structure which has the amorphous luminous layer in which a low-battery drive and high brightness luminescence are possible, research and development of luminescence of R, G, and B in three primary colors, stability, brightness elevation, a laminated structure, the production method, etc. have been done briskly in every direction.

[0016] Furthermore, although it is the feature of an organic material, various new material is invented by the molecular design etc. and the application study to the color display of the organic electroluminescence display device which has the feature which was [luminescence / self-/ a direct-current low-battery drive, a thin shape,] excellent is also beginning to be performed briskly.

[0017] Organic electroluminescence devices (an organic EL element may be called hereafter) have the feature ideal as a spontaneous light [, such as transforming electrical energy into a light energy and emitting light in the shape of a field,] type display device by pouring in current.

[0018] Drawing 12 shows an example of conventional organic EL element 10. This organic EL element 10 produces the ITO (Indium tin oxide) transparent electrode 5, the hole transporting bed 4, a luminous layer 3, the electronic transporting bed 2, and cathode (for example, aluminum electrode) 1 one by one by the vacuum deposition method on the transparent substrate (for example, glass substrate) 6.

[0019] And the thing for which direct current voltage 7 is alternatively impressed between the transparent electrodes 5 and cathode 1 which are an anode plate, The electron into which the hole as a carrier poured in from the transparent electrode 5 was poured from cathode 1 through the hole transporting bed 4 reaches a luminous layer 3 through the electronic transporting bed 2, respectively, the reunion of an electronic-hole arises, the luminescence 8 of predetermined wavelength arises from here, and it can observe from the transparent substrate 6 side.

[0020] In a luminous layer 3, you may use together the anthracene which are other photogenes, naphthalene, a phenanthrene, a pyrene, a chrysene, a perylene, a butadiene, a coumarin, an acridine, a stilbene, etc. The electronic transporting bed 2 can be made to contain the mixture with such fluorescence matter etc.

[0021] Drawing 13 shows another conventional example, in this example, it omits a luminous layer 3, makes the electronic transporting bed 2 contain mixture with the above-mentioned fluorescence matter, and shows organic EL-element 10A constituted so that the luminescence 8 of predetermined wavelength might arise from the interface of the electronic transporting bed 2 and the hole transporting bed 4.

[0022] Drawing 14 shows the example of the above-mentioned organic EL element. That is, although the layered product of each organic layer (the hole transporting bed 4, a luminous layer 3, or electronic transporting bed 2) is allotted between cathode 1 and an anode plate 5, these electrodes are made to cross in the shape of a matrix, and it prepares in the shape of a stripe, and a signal level is impressed to time series by the luminance-signal circuit 40 and the control circuit 41 with a built-in shift register, and it constitutes so that light may be made to emit in much intersection positions (pixel), respectively.

[0023] Therefore, of course, it becomes usable also as a picture reproducer as a display by such composition. In addition, the above-mentioned stripe pattern can be arranged for every color of red (R), green (G), and blue (B), and it can constitute as full color or an object for multicolor.

[0024] Generally, the organic thin film layers 2, 3, and 4 which emit light in the display device which used such an organic EL element, and which consists of two or more pixels are pinched between the transparent electrode 5 and the metal electrode 1, and emit light to a transparent-electrode 5 side.

[0025] However, the field also with the above, organic, still more nearly unsolved EL elements exists.

[0026] That is, luminescence by which RGB three primary colors were stabilized when performing

application to the color display of an organic EL element is indispensable conditions. However, in the organic EL element of RGB three primary colors, since the electronic states which the luminescent material of each color has differ, it is thought that the highest occupancy molecular orbital (HOMO) of luminescent material differs also from minimum non-occupying molecular orbital (LUMO) level.

[0027] This shows that the optimal structure in the organic stacked-volume layer of each color changes with colors, and it is shown that the optimal material for luminous layers which sandwich a luminous layer, such as a hole transporting bed and an electronic transporting bed, differs in each color.

[0028] Moreover, the laminating of two or more layers is carried out in fact, and what is considered as plans, such as an improvement of the luminous efficiency of an organic EL element and reinforcement, is used, although light can be theoretically emitted also by the monolayer. Especially a hole pouring layer (buffer layer for pouring in a charge from an anode to an organic material) and a hole transporting bed (layer for pouring in a charge from a hole pouring layer efficiently to a luminous layer) are indispensable to a highly efficient organic EL element.

[0029] Therefore, if it is going to produce the organic EL element which has the optimized luminous layer of RGB three primary colors, the lamination of a different hole pouring layer, a hole transporting bed, a RGB luminous layer, and an electronic transporting bed is required, and 12 kinds of material is needed only in an organic layer.

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EFFECT OF THE INVENTION

[Function and Effect of the Invention] As mentioned above, in this invention, under a vacuum, the heating dissolution is carried out, the component of an organic layer is pelletized below at sublimation temperature, this pellet is supplied to the container for vacuum evaporation using a supply means, and the aforementioned organic layer is physically formed using this pellet. Therefore, the pelletized material which can also supply surrounding contamination quickly nothing and does not have an impurity by the operation like sublimation refining in the process of pellet production or removal of a remains solvent can be used, without dispersing at the time of supply of the material. Therefore, while the electroluminescence devices which have the organic layer by which the property was stabilized are obtained, material supply can be performed in a vacuum using a pellet supply means, and electroluminescence devices can be manufactured with productivity sufficient at the multi-chamber process.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] However, when it is necessary to produce by the multi-chamber process and application to a color display is performed for the improvement in a property of an organic EL element, considering realizing a low-battery drive by layer structure which is different in each color, an extensive improvement of a facility of the vacuum evaporation system which is production equipment will be needed, and it will become large-sized equipment actually.

[0031] Furthermore, although considering mass-production nature it is required to supply material quickly and simple, the organic material used for organic EL-element production is usually powder in many cases. Therefore, after carrying out air opening of the vacuum evaporation system, the case where exchange the vacuum evaporation boat containing the organic material, or pay [that an organic material does not disperse outside and] careful attention, and it adds directly into a crucible is almost the case.

[0032] However, in the conventional process like the above, it is easy to mix the dust and impurity from the outside, and dispersion and the yield of a property of a product in mass production are affected.

[0033] Therefore, although it is desirable to perform supply of material in the process of vacuum consistency quickly [it is quick and] if it can do without an organic material polluting the circumference in order to solve these problems, the method of effective material supply does not exist.

[0034] Then, the purpose of this invention has supply of material in offering the quick manufacture method of the manufacture method of the high electroluminescence devices of the productivity which can be performed in a vacuum process, without polluting the circumference, its equipment, and the pellet for electroluminescence devices.

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MEANS

[Means for Solving the Problem] That is, an organic layer including a luminescence field faces manufacturing the electroluminescence devices by which the laminating is carried out, and pelletizes the component of the aforementioned organic layer, and this invention requires it for the manufacture method (the manufacture method of this invention is called hereafter.) of electroluminescence devices which forms the aforementioned organic layer physically using this pellet.

[0036] The pelletized material which can also supply surrounding contamination quickly nothing and does not have an impurity by the operation like sublimation refining in the process of pellet production or removal of a remains solvent can be used without dispersing at the time of the supply, since an organic layer is formed using the pelletized component according to the manufacture method of this invention. Therefore, while the electroluminescence devices which have the organic layer by which the property was stabilized are obtained, material supply can be performed in a vacuum using a pellet supply means, and electroluminescence devices can be manufactured with productivity sufficient at the multi-chamber process.

[0037] Moreover, this invention requires an organic layer including a luminescence field for the manufacturing installation (the manufacturing installation of this invention is called hereafter.) of the electroluminescence devices characterized by having a container, the electrode holder which holds the pellet of the component as the aforementioned organic layer, and a supply means to supply the aforementioned pellet held in this electrode holder in the aforementioned container in the equipment which manufactures the electroluminescence devices by which the laminating is carried out.

[0038] According to the manufacturing installation of this invention, a pellet can be held in an electrode holder, a container can be supplied, and a manufacturing installation with the sufficient repeatability of the above-mentioned manufacture method can be offered.

[0039] Moreover, an organic layer including a luminescence field faces this invention manufacturing the pellet used for the membrane formation of the aforementioned organic layer of electroluminescence devices by which the laminating is carried out, and it requires it for the manufacture method (the manufacture method of the pellet of this invention is called hereafter.) of a pellet which carries out the heating dissolution and pelletizes the raw material of the component of the aforementioned organic layer below at sublimation temperature under a vacuum.

[0040] According to the manufacture method of the pellet of this invention, since the heating dissolution of the raw material is carried out below at sublimation temperature under a vacuum, sublimation refining of the raw material is carried out, a remains solvent is removed in this process, and the manufacture method of the pellet of the organic material for electroluminescence devices which has the organic layer which consists of a pure material without a pollutant can be offered.

[0041] here -- setting -- "a vacuum" -- 10^{-3} Torr (0.13Pa) -- further -- the state of a high vacuum is pointed out from 10^{-6} Torr (1.3×10^{-4} Pa), or it

[0042]

[Embodiments of the Invention] Hereafter, the gestalt of desirable operation of this invention is explained, referring to a drawing.

[0043] In the manufacture method of the manufacture method of the above-mentioned this invention, a manufacturing installation, and a pellet, although it is desirable to use a pellet with a size of 1mm - several cm which processed the raw material of an organic material as the aforementioned pellet, size is not limited to this.

[0044] It is desirable to put into a container with an open sand mold or a lid as shown in drawing 3 , to carry out the heating dissolution, to anneal the raw material of the aforementioned component below at sublimation temperature under a vacuum, in production of this pellet, and to produce the aforementioned pellet. Thereby, since a large material of sublimability adheres to the internal surface of a top cover 33 and a small material of sublimability melts and stops in the dissolution pan 31 where sublimation refining of the raw material was carried out and a remains impurity is removed, as shown in drawing 4 , this container 30 can be disassembled and pellets can be collected easily.

[0045] Therefore, aluminum quinoline complex Alq₃ which a raw material shows to drawing 8 When it is desirable to use a container with a lid when sublimability is large like alpha-NPD shown in drawing 7 and sublimability is small like m-MTDATA which a raw material shows to drawing 6 , the container of an open sand mold can be used.

[0046] Moreover, things other than this are sufficient as a container, and it can devise internal structure and can form it arbitrarily so that the pellet of a fixed configuration may be obtained.

[0047] And as typically shown in drawing 1 , it is desirable to put the produced pellet into the container 23 for vacuum evaporatio for every component kind of the aforementioned organic layer, to convey the substrate 24 in which ITO was formed within a chamber 20, and to carry out vacuum deposition one by one.

[0048] In this case, as shown in drawing 2 , in order to hold a pellet in an electrode holder 21, to carry out natural fall of the aforementioned pellet into the aforementioned container 23 for vacuum evaporatio from this electrode holder 21 and to form the specified quantity, it is desirable to supply the material of a complement.

[0049] By forming an organic layer using the pellet obtained as mentioned above, optically, on a transparent base, the laminating of a transparent electrode, an organic hole transporting bed, an organic luminous layer and/or an organic electronic transporting bed, and the metal electrode can be carried out one by one optically, and it can constitute as good organic electroluminescence devices.

[0050] Moreover, the organic EL element to produce may carry out the laminating of the ITO anode electrode 12, the hole pouring layer 13, the hole transporting bed 14, a luminous layer 15, the electronic transporting bed 16, and the cathode electrode 17 on a substrate 11, as shown in drawing 5 , and especially this component is not limited. For example, if it is the hole transporting bed 14, hole transportation material, such as a benzidine derivative, a styryl amine derivative, a triphenylmethane-color derivative, and a hydrazone derivative, can also be used. Moreover, if the electronic transporting bed 16 is made to contain a photogene like drawing 13 mentioned already, and it can also be made the single hetero type with which the electronic transporting bed 16 made the luminous layer serve a double purpose and the metal electrode of a cathode is formed thinly, luminescence can also be taken out from a cathode side.

[0051] Moreover, the ITO anode electrode 12, the hole pouring layer 13, the hole transporting bed 14, a luminous layer 15, the electronic transporting bed 16, and the cathode electrode 17 may be laminated structures which each becomes from two or more layers.

[0052] Furthermore, vapor codeposition of a minute amount molecule may be performed for control of the emission spectrum of a luminous layer 15, for example, you may be a minute amount **** organic thin film about organic substances, such as a perylene derivative, a coumarin derivative, and pyran system coloring matter.

[0053] About the material of the cathode electrode 17, in order to pour in an electron efficiently, it is desirable to use the small metal of the work function from the vacuum level of an electrode material, it is a simple substance about low work function metals, such as In, Mg, Ag, calcium, Ba, and Li, or you may use it as an alloy with other metals, raising stability.

[0054] In the gestalt of this operation, although ITO which is a transparent electrode is used for the

anode electrode 12 in order to take out luminescence from an anode electrode side, in order to pour in a hole efficiently, of course, you may use electrodes, such as what has a large work function from the vacuum level of an anode electrode material, for example, Au, SnO_2+Sb , and $\text{ZnO}+\text{aluminum}$.

[0055] Furthermore, in order to raise stability, as an imaginary line shows, a protective coat may be prepared in drawing 5 with a germanium oxide etc., the influence of the oxygen in a closure deed and the atmosphere etc. may be eliminated, and an element may be driven in the state where it lengthened to the vacuum, of course.

[0056] Moreover, heat-treatment, sublimation processing, and an organic solvent may be used for the method of pelletizing, pellet size needs to be influenced by a property, a pelletizing art, etc. of material, in a small thing, although it becomes the shape of a several centimeters pellet from 1mm by the big thing, pellet size does not need to be single, for example, the several cm pellet may be intermingled from 1mm.

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EXAMPLE

[Example] Hereafter, the example of this invention is explained.

[0058] According to the gestalt of desirable operation of this invention mentioned above, membrane formation equipment (a chamber may be called hereafter) was produced. Drawing 1 is the ** type view showing the composition superficially, and drawing 2 is some of these outline cross sections.

[0059] A pellet 25 is put into a crucible 26 for every material which constitutes an organic layer, it melts, and the number of material kinds required as a fusion material 26 is allotted, as shown in drawing 2, the electrode holder 21 which put in the pellet 25 for supply on the crucible 26 further is arranged, and the supply races 22 are formed successively at the nose of cam of an electrode holder 21, and it is formed in the configuration which is easy to supply a pellet to a crucible 26.

[0060] Drawing 1 shows that membranes are formed one by one and an organic layer is taken out to Arrow out side, when the state where 3 sets has been placed in a fixed position in one vacuum chamber (illustration ellipsis) in the group of such an electrode holder 21 and a crucible 23 is shown, and it is carried in in a chamber, and the substrate 24 after forming an ITO transparent electrode from Arrow in side has the crucible 23 top for every material kind conveyed and goes round it.

[0061] A crucible 23 is supplied from an electrode holder 21 at each using the natural fall by gravity. The timing of supply establishes a calibration curve from the relation between the heating temperature (the amount of current supplied) of the crucible 23 according to each organic material, and an evaporation rate, the decrement of the material in a crucible 23 is estimated, and the feedback system is taken so that the melting material 26 may always exist in a crucible 23.

[0062] Drawing 3 shows the container for producing a pellet, (a) is an outline cross section, (b) is a plan, and drawing 4 shows the cross section in the state where this container was disassembled.

[0063] Like illustration, this container 30 consists of dissolution **** 31, an inside lid 32, and a top cover 33, and Collars 31a, 32a, and 33a are installed, and if these collars 31a, 32a, and 33a pile up, it has structure which can carry out a seal from each rim. As a material of a container 30, a tantalum, molybdenum, or a tungsten can use it suitably.

[0064] The imaginary line shown in drawing 3 (a) and drawing 4 shows the pellet 25 under production. As shown in drawing 3 (a), the raw material 27 with large sublimability is paid into dissolution **** 31, closes the inside lid 32 and a top cover 33, and heats them by resistance heating.

[0065] Like illustration, the hole 34 is established in the inside [this] lid 32 at two places, and when sublimability is a large raw material, it is used in the state of drawing 3 (a). Therefore, once it heats and sublimates the heated large raw material of sublimability and it hits a top cover 33 from this hole 34, it comes out to the hole 35 shell exterior of a top cover 33. However, since it heats near the sublimation temperature, there are few amounts which come out to the exterior, and many adhere to internal-surface 33b of a top cover 33.

[0066] However, it is **** to the ***** dissolution pan 31, without sublimating, since it heats below at sublimation temperature, when sublimability is a small raw material. Therefore, only using the dissolution pan 31, even if the inside lid 32 and a top cover 33 do not use it, they can make a pellet.

[0067] Each production of the above-mentioned pellet is performed within the vacuum system

(illustration ellipsis) according to a vacuum chamber or it. And drawing of the produced pellet can be easily taken out, if a container 30 is disassembled as shown in drawing 4 . Although there is no limit in the degree of vacuum of pellet production, the degree of high vacuum can obtain the pellet of a high grade preferably.

[0068] Drawing 5 shows an example of the organic EL element which formed the pellet 25 mentioned above as a material of an organic layer, and carries out the laminating of the metal electrode 17 as the ITO transparent electrode 12, the hole pouring layer 13, the hole transporting bed 14, the luminous layer 15, the electronic transporting bed 16, and cathode as an anode one by one on a glass substrate 11.

[0069] And while forming the organic EL element using the pellet produced from the raw material like the example shown below as a component of the organic layer of this organic EL element, the organic EL element using the powdery raw material which is not pelletized was formed as an example of comparison, and both were compared.

[0070] <Example 1> m-MTDATA of the structure expression shown in drawing 6 as a hole-injection layer material in this example (4, 4', 4''-tris(3-methylphenylphenylamino) triphenylamine) It used. As shown in pelletizing processing at drawing 3 , 1.0g of m-MTDATA was put into the dissolution pan 11 made from an open-sand-mold tantalum (the same is said of each following example) which uses neither a top cover nor an inside lid, and it became hot slowly until material dissolved in the vacuum. Dissolved m-MTDATA became a vitrified pellet, before the sublimation after dissolution occurred, it stopped heating, and it annealed it to the room temperature.

[0071] The existence of sublimation checked by the thickness sensor monitor. Pelletizing of this m-MTDATA was simultaneously performed using four containers 30 shown in drawing 3 . Thus, the produced m-MTDATA pellet turned into a pellet with a 20mmx10mmx thickness of about 5mm.

[0072] <Example 2> In this example, alpha-NPD (4 and 4'-bis[N-(1-naphthyl)-N-phenyl-amino] biphenyl) of the structure expression shown in drawing 7 was used as an electron hole transporting-bed material. Compared with m-MTDATA, sublimability of alpha-NPD is large, and it is difficult to pelletize by sublimation of material only in an open-sand-mold dissolution pan 11 like an example 1. Therefore, as pelletizing processing, as shown in drawing 3 , it carried out with the lid. 1.0g of alpha-NPD was put in into the state of drawing 3 , and in the vacuum, material became hot slowly until sublimation had been sensed by the thickness sensor monitor. And in the 1/100nm or less state, the evaporation rate stopped heating and cooled slowly to the room temperature.

[0073] Pelletizing of this alpha-NPD was simultaneously performed using four containers 30 like the example 1. Thus, the produced alpha-NPD pellet turned into a pellet with a 15mmx10mmx thickness of about 3mm, adhered to internal-surface 33b of a top cover 33, and was pelletized.

[0074] <Example 3> Alq3 of the structure expression shown in drawing 8 as an electronic transportability luminous layer material in this example (8-hydroxy quinorine aluminum) It used. Alq3 Compared with m-MDATA, sublimability is large, and pelletizing is difficult by sublimation of material like an example 2 in the dissolution pan 11 of an open sand mold. Therefore, it uses with a lid like an example 2 as pelletizing processing, and is Alq3 similarly. 1.0g was put in, and in the vacuum, material became hot slowly until sublimation had been sensed by the thickness sensor monitor. And in the state 1/100nm or less, the evaporation rate stopped heating and cooled slowly to the room temperature.

[0075] This Alq3 Pelletizing was simultaneously performed using four containers 30 like the example 1. Thus, produced Alq3 The pellet turned into a pellet with a 15mmx10mmx thickness of about 3mm, adhered to internal-surface 33b of a top cover 33, and was pelletized.

[0076] <Example 4> In this example, an organic EL element like drawing 5 was produced using the pellet produced by the above-mentioned examples 1-3. As shown in drawing 10 , size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx glass substrate 11 which is 30mm SiO2 The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. It is Alq3 considering alpha-NPD as 20nm and an electronic transporting bed 16 considering m-MTDATA as 30nm and a hole transporting bed 14 as a hole pouring layer 13 on this substrate. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation

of aluminum was performed as a metal electrode 17 of a cathode.

[0077] Using the vacuum evaporation system 20 shown in drawing 1, in early stages of vacuum evaporation, beforehand, production of this organic EL element put one pellet 25 of each material at a time into the crucible 23, and set it to it. As mentioned already after that, the substrate of a 30mm angle like drawing 10 was able to be produced at about 200 time [1] by supplying a pellet 25, without carrying out air opening of the chamber 20.

[0078] <Example 5> BSB-BCN of the structure expression shown in drawing 9 as a luminescent material in this example (2, 5-Bis[4-(N and N-4-methoxydiphenylamino) styryl]-1, 4-dicyanobenzene) It used. 1.0g of BSB-BCN was put into the dissolution pan 11 of an open sand mold like the example 1 as pelletizing processing, and it became hot slowly until material dissolved in the vacuum. Dissolved BSB-BCN became a vitrified pellet, before the sublimation after dissolution occurred, it stopped heating, and it annealed it to the room temperature.

[0079] The existence of sublimation checked by the thickness sensor monitor. Pelletizing of this BSB-BCN was simultaneously performed like the above-mentioned examples 1-3 using four containers 30 shown in drawing 3. Thus, the produced m-MTDATA pellet turned into a pellet with a 20mmx10mmx thickness of about 5mm.

[0080] Next, the organic EL element which emits light in red like drawing 5, using the pellet which consists of this BSB-BCN as a luminous layer was produced. As shown in drawing 10, size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx glass substrate 11 which is 30mm SiO₂ The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. m-MTDATA as a hole pouring layer 13 on this substrate 30nm, BSB-BCN as 30nm and a luminous layer 15 for alpha-NPD as a hole transporting bed 14 30nm, It is Alq₃ as an electronic transporting bed 16. The laminating of the 30nm was carried out one by one, and, finally the vacuum evaporation of AlLi (aluminum lithium alloy and Li concentration % of one mol) was performed as a metal electrode 17 of a cathode.

[0081] Using the vacuum evaporation system 20 shown in drawing 1 like the example 4, in early stages of vacuum evaporation, production of this organic EL element put one pellet 25 of each material at a time into the crucible 3 beforehand, and set it to it. As mentioned already after that, about 200 substrates of a 30mm angle like drawing 10 were able to be produced at once by supplying a pellet 25, without carrying out air opening of the chamber 20.

[0082] <Example 1 of comparison> In this example, it did not pelletize but an organic EL element like drawing 5 (the same is said of each following example of comparison) was produced using each organic powdered layer material. As shown in drawing 10, size L namely, on the ITO substrate which formed the ITO electrode 12 (about 100nm of thickness) on the 30mmx glass substrate 11 which is 30mm SiO₂ The ITO glass substrate for organic EL-element production which carried out the mask of except for the luminescence field whose size l is 2mmx2mm by vacuum evaporation is produced. Powdered m-MTDATA, alpha-NPD, and Alq₃ It is made to correspond to the examples 1-3 which mentioned each material already. It puts in about g. the container of drawing 3 -- 0.5-0.7 -- m-MTDATA 30nm as a hole pouring layer 13 on this substrate It is Alq₃ considering alpha-NPD as 20nm and an electronic transporting bed 16 as a hole transporting bed 14. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation of aluminum as a metal electrode 17 of a cathode was performed.

[0083] However, by this method, by the time material was lost, the limitation carried out the vacuum evaporation of the about 20-30 substrates of a 30mm angle like drawing 10.

[0084] <Example 2 of comparison> It is Alq₃ considering alpha-NPD as 20nm and an electronic transporting bed 16 considering m-MTDATA as 30nm and a hole transporting bed 14 as a hole pouring layer 13 on the substrate produced like the above-mentioned example 1 of comparison in this example. The laminating of the 50nm was carried out one by one, and, finally the vacuum evaporation of aluminum was performed as a metal electrode 17 of a cathode. By this method, although about 40 substrates production was [***** / before supplying material] possible, there was 20-30 number of

sheets which can produce after supply of material.

[0085] Therefore, in order to pursue this cause, when the material distribution system was checked, the powdered organic material adhered to the rail 22 for material supply shown in drawing 1 and drawing 2, and it remained, without falling into a crucible 23 completely. Moreover, the organic material might fall around. Furthermore, as compared with the device of an example 4, luminous efficiency was low, mixing of material etc. was suggested and the organic EL element produced after material supply had a bad yield.

[0086] <Example 3 of comparison> BSB-BCN of the structure expression shown in drawing 9 as a luminescent material in this example (2, 5-Bis[4-(N and N-4-methoxydiphenylamino) styryl]-1, 4-dicyanobenzene) Although used, it did not pelletize, but raw material was supplied in the state of powder, and the organic EL element of red luminescence was produced.

[0087] That is, it considers as the hole pouring layer 13 on the substrate produced like the above-mentioned example of comparison, and is Alq3 considering BSB-BCN as 30nm and an electronic transporting bed 16 considering alpha-NPD as 30nm and a luminous layer 15 considering m-MTDATA as 30nm and a hole transporting bed 14. The laminating of the 30nm was carried out one by one, and, finally the vacuum evaporation of AlLi (aluminum lithium alloy and Li concentration % of one mol) was performed as a metal electrode 17 of a cathode.

[0088] Drawing 11 is the organic EL element shown in drawing 5, and is the graph of the data which compare the brightness and luminous efficiency the case where it uses with the material which pelletized BSB-BCN of an example 5 as a luminous layer 15, and at the time of using the powder material of BSB-BCN shown in the example 3 of comparison as a luminous layer 15.

[0089] As shown in this graph, when it deposits with the material which pelletized BSB-BCN Brightness is about 8000 cd/m² at the time of applied-voltage 10.5V. When it is shown and deposits by powdered BSB-BCN to the luminous efficiency at this time being between 0.45 - 0.5 lm/W, brightness is cd [8000]/m² at 11V. Although it is almost the same Since luminous efficiency has become between 0.25 - 0.3 lm/W, in the case of a powdered material, it turns out that the property of an element is clearly inferior. That is, by pelletizing material, moisture, an impurity, etc. of a raw material are removed and it can be proved that it is an effect by the purity of material having been improved.

[0090] the material kind which constitutes the organic layer of an organic EL element according to this example -- since vacuum evaporation can be performed supplying the material moreover pelletized to the crucible 23 in the vacuum using the material pelletized to things, air opening of the vacuum evaporation chamber cannot be carried out because of vacuum evaporation material supply, and a lot of elements can be produced at once

[0091] Moreover, since the container for pellet production can be used irrespective of the size of the sublimability of material, it is efficient, and since it can decompose, drawing of the produced pellet is also easy.

[0092] Furthermore, by being also the head end process of vacuum evaporation material, consequently performing sublimation refining and removal of a remains solvent simultaneously, and using this pellet, the process of pelletizing is stabilized and, in other words, can produce the efficient device by which the property was stabilized with the sufficient yield.

[0093] The above-mentioned example can be deformed based on the technical thought of this invention.

[0094] For example, the vacuum evaporation equipment shown in drawing 1 can also be made into the composition and the configurations other than an example, and can be carried out arbitrarily [the supply method of the pellet of a crucible 23 shown in drawing 2] besides an example.

[0095] Moreover, the container 30 for pellet production shown in drawing 3 can also be made into the composition and the configurations other than an example. In order to standardize the configuration of the pellet produced especially in a fixed configuration, a diaphragm can also be formed in the internal surface of dissolution **** 31 and a top cover 33.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the ** type view of the vacuum evaporation system by the example of this invention.

[Drawing 2] It is the outline cross section showing a part of **** vacuum evaporation system.

[Drawing 3] The pellet production container by the **** example is shown, (a) is an outline cross section and (b) is a plan.

[Drawing 4] It is the exploded view of a **** pellet production container.

[Drawing 5] It is the outline cross section of the organic EL element by the **** example.

[Drawing 6] It is the structure expression of m-MTDATA used for the **** organic EL element.

[Drawing 7] It is the structure expression of alpha-NPD used for the **** organic EL element.

[Drawing 8] Alq3 used for the **** organic EL element It is a structure expression.

[Drawing 9] It is the structure expression of BSB-BCN used for the **** organic EL element.

[Drawing 10] It is the plan of the morphosis of a **** organic EL element.

[Drawing 11] It is the graph which compared and showed the property of a **** organic EL element.

[Drawing 12] It is the outline cross section showing the organic EL element by the conventional example.

[Drawing 13] Other examples of a **** organic EL element are *(ed). It is an outline cross section.

[Drawing 14] It is the outline cross section showing the example of a **** organic EL element.

[Description of Notations]

11 [-- Hole pouring layer,] -- A substrate, 12 -- An ITO electrode (anode), 13 14 [-- An electronic transporting bed, 17 / -- Metal electrode (cathode),] -- A hole transporting bed, 15 -- A luminous layer, 16 20 [-- A supply rail, 23 / -- A crucible, 24 / -- An ITO substrate, 25 / -- A pellet, 26 / -- Fusion material, 27 / -- A raw material, 30 / -- A container, 31 / -- A dissolution pan,, 31a, 32a, 33a / -- A collar, 32 / -- An inside lid, 33 / -- A top cover, 33b / -- An internal surface, 34 / -- Hole] -- A vacuum evaporation system, 21 -- An electrode holder, 22

[Translation done.]

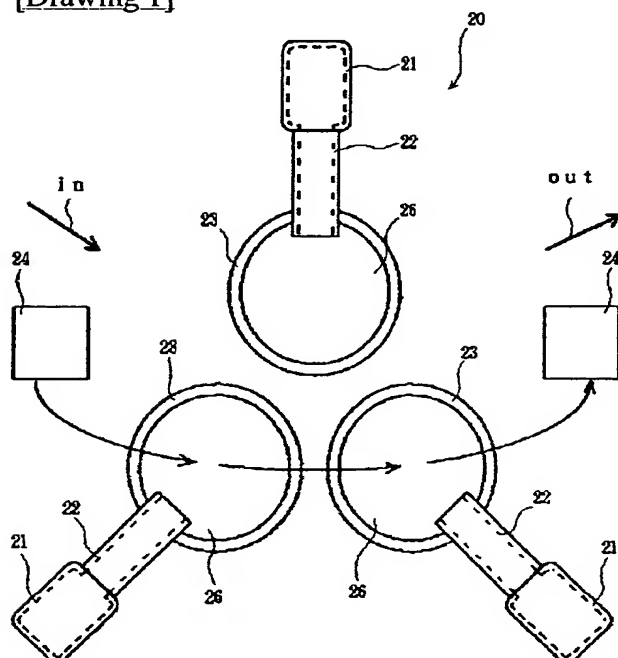
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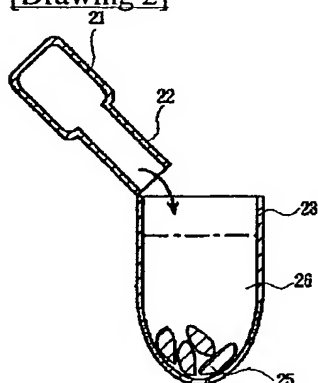
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DRAWINGS

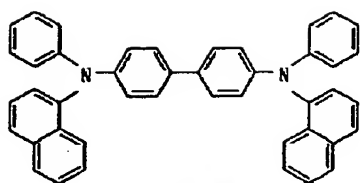
[Drawing 1]



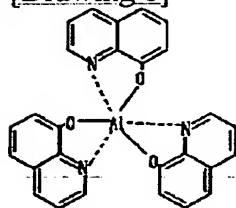
[Drawing 2]



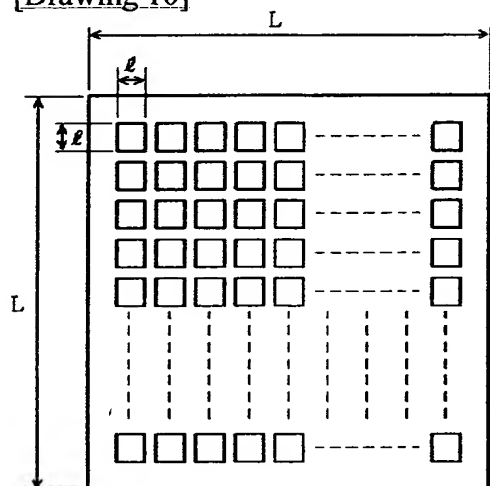
[Drawing 3]

 α -NPDの構造

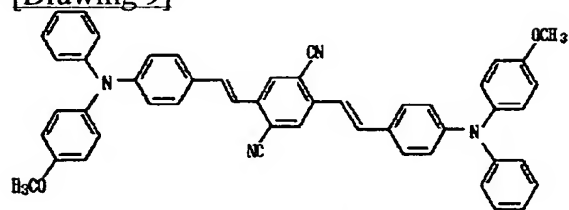
[Drawing 8]

Alq₃の構造

[Drawing 10]

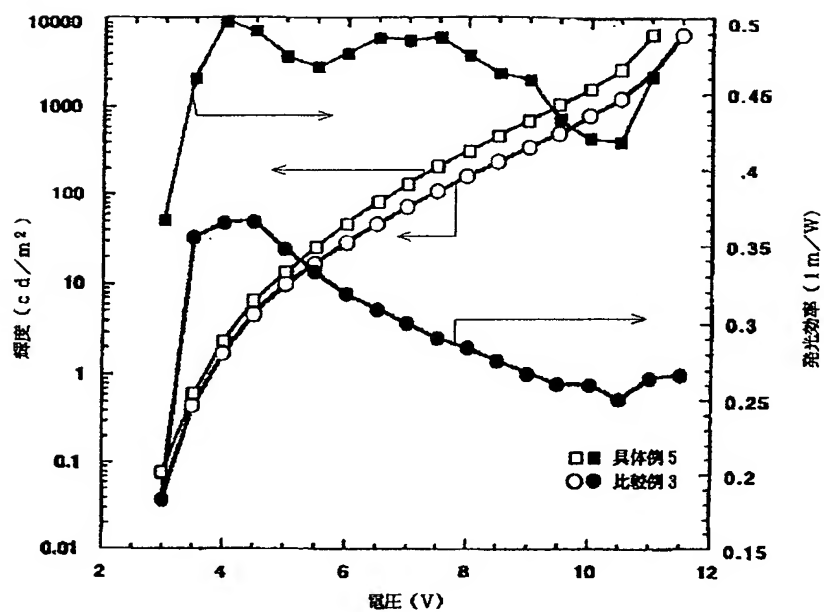


[Drawing 9]

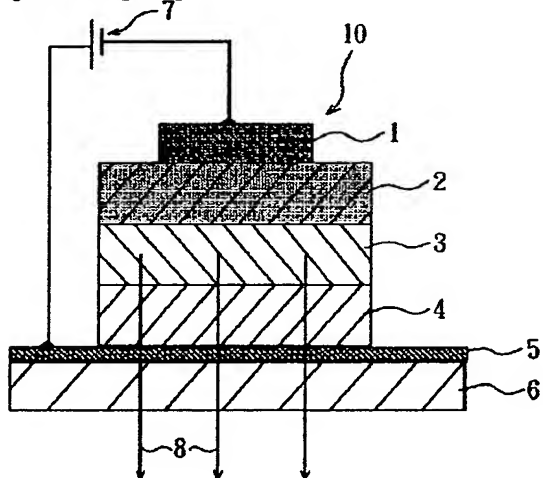


BSB-BCNの構造

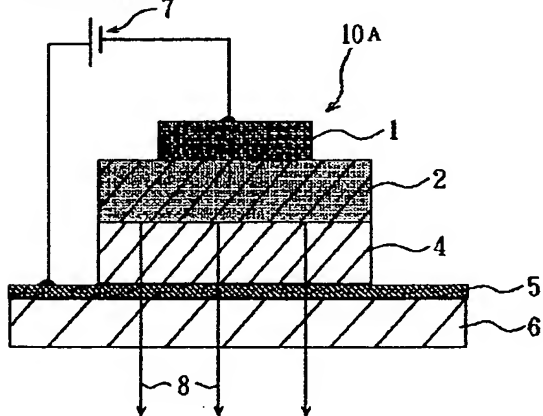
[Drawing 11]



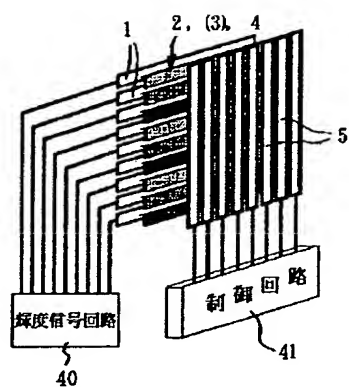
[Drawing 12]



[Drawing 13]



[Drawing 14]



[Translation done.]